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14. ABSTRACT
This research project theoretically exploited the unique properties of nanoscale graphene based structures for highly functional spintronic applications at room temperature. To overcome the non-magnetic nature of intrinsic graphene, two promising phenomena were explored that can introduce desired spin magnetic functionalities with electrical control. The first approach incorporates the magnetism by forming a hybrid structure with appropriate magnetic materials. The interactions between graphene electrons and magnetic ions at the interfaces result in

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Report Title

Final Technical Report: Graphene Nanostructures for Novel Spin Magnetic Device Applications

ABSTRACT

This research project theoretically exploited the unique properties of nanoscale graphene based structures for highly functional spintronic applications at room temperature. To overcome the non-magnetic nature of intrinsic graphene, two promising phenomena were explored that can introduce desired spin magnetic functionalities with electrical control. The first approach incorporates the magnetism by forming a hybrid structure with appropriate magnetic materials. The interactions between graphene electrons and magnetic ions at the interfaces result in effective magnetic fields that can affect the characteristics of both graphene and magnetic layers. The second approach introduces the magnetic effects by utilizing non-zero magnetic moments induced at the edge states, defects, vacancies, etc. Both of these effects may be readily amenable for electrical control through the dependence on graphene electronic properties. With the development of appropriate models, the main focus of investigation was to analyze the physical properties and basic functionalization principles, particularly as the dimension of the structure shrinks, and to examine their device potential including spin magnetic switches. As an extension of the original concepts, the research project also examined spin magnetic properties of topological insulators and their potential applications; with strong spin-orbit coupling, this emerging material system with nanoscale 2D electron states offers a new opportunity in a manner analogous to the graphene based hybrid structures.

Enter List of papers submitted or published that acknowledge ARO support from the start of the project to the date of this printing. List the papers, including journal references, in the following categories:

(a) Papers published in peer-reviewed journals (N/A for none)

Received		Paper
08/16/2011	2.00	J. Mullen, K. Borysenko, X. Li, Y. Semenov, J. Zavada, M. Nardelli, K. Kim. Electron-phonon interactions in bilayer graphene, Physical Review B, (04 2011): 0. doi: 10.1103/PhysRevB.83.161402
08/16/2011	7.00	X. Li, E. A. Barry, J. M. Zavada, M. Buongiorno Nardelli, K. W. Kim. Influence of electron-electron scattering on transport characteristics in monolayer graphene, Applied Physics Letters, (08 2010): 0. doi: 10.1063/1.3483612
08/16/2011	6.00	Y Chen, T Jayasekera, A Calzolari, K W Kim, M Buongiorno Nardelli. Thermoelectric properties of graphene nanoribbons, junctions and superlattices, Journal of Physics: Condensed Matter, (09 2010): 0. doi: 10.1088/0953-8984/22/37/372202
08/16/2011	3.00	X. Li, E. A. Barry, J. M. Zavada, M. Buongiorno Nardelli, K. W. Kim. Surface polar phonon dominated electron transport in graphene, Applied Physics Letters, (12 2010): 0. doi: 10.1063/1.3525606
08/16/2011	4.00	Y. Semenov, J. Zavada, K. Kim. Electron spin relaxation in carbon nanotubes, Physical Review B, (10 2010): 0. doi: 10.1103/PhysRevB.82.155449
12/11/2012	5.00	V. A. Kochelap, K. W. Kim, V. N. Sokolov. Magnetoconcentration effect in intrinsic graphene ribbons, Applied Physics Letters, (09 2010): 112112. doi: 10.1063/1.3486124
12/11/2012	15.00	Y. G. Semenov, J. M. Zavada, K. W. Kim. Graphene spin capacitor for magnetic field sensing, Applied Physics Letters, (07 2010): 13106. doi: 10.1063/1.3462297
12/11/2012	14.00	Yuriy Semenov, Xiaodong Li, Ki Kim. Tunable photogalvanic effect on topological insulator surfaces via proximity interactions, Physical Review B, (11 2012): 201401. doi: 10.1103/PhysRevB.86.201401
12/11/2012	16.00	Thushari Jayasekera, B. D. Kong, K. W. Kim, M. Buongiorno Nardelli. Band Engineering and Magnetic Doping of Epitaxial Graphene on SiC (0001), Physical Review Letters, (04 2010): 146801. doi: 10.1103/PhysRevLett.104.146801
12/11/2012	17.00	Y. G. Semenov, J. M. Zavada, K. W. Kim. Electrically controlled magnetic switching based on graphene-magnet composite structures, Journal of Applied Physics, (03 2010): 64507. doi: 10.1063/1.3354066
12/11/2012	8.00	Y. Semenov, J. Zavada, K. Kim. Weak ferromagnetism of antiferromagnetic domains in graphene with defects, Physical Review B, (10 2011): 165435. doi: 10.1103/PhysRevB.84.165435
12/11/2012	9.00	K. Borysenko, M. Buongiorno Nardelli, K. Kim, X. Li. Electron transport properties of bilayer graphene, Physical Review B, (11 2011): 195453. doi: 10.1103/PhysRevB.84.195453
12/11/2012	10.00	X. Li, B. D. Kong, J. M. Zavada, K. W. Kim. Strong substrate effects of Joule heating in graphene electronics, Applied Physics Letters, (12 2011): 233114. doi: 10.1063/1.3668113
12/11/2012	11.00	M. Buongiorno Nardelli, R. Mao, B. D. Kong, K. W. Kim, T. Jayasekera, A. Calzolari. Phonon engineering in nanostructures: Controlling interfacial thermal resistance in multilayer-graphene/dielectric heterojunctions, Applied Physics Letters, (09 2012): 113111. doi: 10.1063/1.4752437
12/11/2012	12.00	X. Duan, V. A. Stephanovich, Y. G. Semenov, K. W. Kim. Magnetic domain wall transfer via graphene mediated electrostatic control, Applied Physics Letters, (07 2012): 13103. doi: 10.1063/1.4732794

12/11/2012	13.00	Yuriy Semenov, Xiaopeng Duan, Ki Kim. Electrically controlled magnetization in ferromagnet-topological insulator heterostructures, Physical Review B, (10 2012): 161406. doi: 10.1103/PhysRevB.86.161406

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Number of Papers published in peer-reviewed journals:

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Received		Paper
08/16/2011	1.00	B. D. Kong, Y. G. Semenov, C. M. Krowne, K. W. Kim. Unusual magnetoresistance in a topological insulator with a single ferromagnetic barrier, Applied Physics Letters, (06 2011): 243112. doi: 10.1063/1.3600330

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Number of Papers published in non peer-reviewed journals:

(c) Presentations

E. A. Barry, J. T. Mullen, K. Borysenko, T. Jayasekera, B. Kong, K. W. Kim, and M. Buongiorno Nardelli, "First Principles Analysis of Electron-Phonon Scattering Rates in Graphene and Their Influence on Electronic Transport," presented at the Army Research laboratory Workshop on Graphene Electronics (August, 2009. Adelphi, Maryland).

B. D. Kong, T. Jayasekera, K. W. Kim, and M. Buongiorno Nardelli, "Band Engineering and Magnetic Doping of Epitaxial Graphene on SiC," presented at the March Meeting of the American Physical Society (March, 2010, Portland, Oregon), Bull. Am. Phys. Soc. 55, H22.00005 (2010).

Y. F. Chen, T. Jayasekera, B. D. Kong, K. W. Kim, and M. Buongiorno Nardelli, "Thermal Transport in Graphitic Nanostructures: Analytic Force Constants and First Principles Calculations," presented at the March Meeting of the American Physical Society (March, 2010, Portland, Oregon), Bull. Am. Phys. Soc. 55, P21.00010 (2010).

Y. G. Semenov, J. M. Zavada, K. W. Kim, "Electron Spin Relaxation in Carbon Nanotubes: Dyakonov-Perel Mechanism," presented at the March Meeting of the American Physical Society (March, 2010, Portland, Oregon), Bull. Am. Phys. Soc. 55, V35.00013 (2010).

T. Jayasekera, B. D. Kong, K. W. Kim, and M. Buongiorno Nardelli, "Band Engineering and Magnetic Doping of Epitaxial Graphene on SiC (0001)," presented at 2010 Graphene Week (April, 2010, College Park, Maryland).

E. A. Barry, X. Li, J. M. Zavada, M. Buongiorno Nardelli, and K. W. Kim, "Influence of Electron-Electron Scattering and Substrate Effects on the Transport Properties of Monolayer Graphene," presented at 2010 Graphene Week (April, 2010, College Park, Maryland).

Y. G. Semenov, J. M. Zavada, and K. W. Kim, "Electron Spin Relaxation in Carbon Nanotubes," presented at the Third International World University Network (WUN) Conference on Spintronic Materials and Devices (Urbana, Illinois, June, 2010).

Y. G. Semenov, J. M. Zavada, and K. W. Kim, "Efficient Electron Spin Relaxation via Dyakonov-Perel Type Mechanism in Carbon Nanotubes," presented at the 55th Magnetism and Magnetic Materials Conference (November, 2010, Atlanta, Georgia).

Y. G. Semenov, J. M. Zavada, and K. W. Kim, "Magnetic Correlation in Graphene with Vacancies," presented at the 55th Magnetism and Magnetic Materials Conference (November, 2010, Atlanta, Georgia).

K. M. Borysenko, J. T. Mullen, X. Li, Y. G. Semenov, J. M. Zavada, M. Buongiorno Nardelli, and K. W. Kim, "Electron Transport Properties of Bilayer Graphene," presented at the March Meeting of the American Physical Society (March, 2011, Dallas, Texas), Bull. Am. Phys. Soc. 56, Q37.00010 (2011).

Y. G. Semenov, J. M. Zavada, K. Borysenko, and K. W. Kim, "Effects of Magnetic Correlation of Localized Spins in Graphene," presented at the March Meeting of the American Physical Society (March, 2011, Dallas, Texas), Bull. Am. Phys. Soc. 56, K1.00173 (2011).

T. Jayasekera, S. Xu, K. W. Kim, and M. Buongiorno Nardelli, "Electronic Properties of the Graphene/SiC (000) Interface: a First Principles Study," presented at the March Meeting of the American Physical Society (March, 2011, Dallas, Texas), Bull. Am. Phys. Soc. 56, P36.00001 (2011).

X. Duan, V. A. Stephanovich, Y. G. Semenov, and K. W. Kim, "Currentless Domain Wall Motion along Biased Ferromagnet/Semiconductor Heterostructures," presented at the 56th Magnetism and Magnetic Materials Conference (November, 2011, Scottsdale, Arizona).

Y. G. Semenov and K. W. Kim, "Magnetoelectric Effect in Topological Insulator/Magnetic Layer Nanostructure," presented at the March Meeting of the American Physical Society (March, 2012, Boston, Mass.), Bull. Am. Phys. Soc. 57, V37.00003 (2012).

R. Mao, B. D. Kong, T. Jayasekera, M. Buongiorno Nardelli, "Nanoscale Thermal Transport in Graphene Interfaces," presented at the March Meeting of the American Physical Society (February, 2012, Boston, Mass.), Bull. Am. Phys. Soc. 57, B6.00001 (2012).

X. Duan, V. A. Stephanovich, Y. G. Semenov, H. Fangohr, M. Franchin, and K. W. Kim, "Electric Field Driven Domain Wall Transfer in Hybrid Structures," presented at the 70th Annual Device Research Conference (June, 2012, University Park, Pennsylvania).

Number of Presentations: 16.00

Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

Received Paper

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Peer-Reviewed Conference Proceeding publications (other than abstracts):

Received Paper

TOTAL:

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(d) Manuscripts

Received Paper

TOTAL:

Number of Manuscripts:

Books

Received Paper

TOTAL:

Patents Submitted

Patents Awarded

Awards

Graduate Students

NAME	PERCENT_SUPPORTED	Discipline
Xiaodong Li	0.15	
Xiaopeng Duan	0.15	
FTE Equivalent:	**0.30**	
Total Number:	**2**	

Names of Post Doctorates

NAME	PERCENT_SUPPORTED
Yuriy Semenov	0.50
FTE Equivalent:	**0.50**
Total Number:	**1**

Names of Faculty Supported

NAME	PERCENT_SUPPORTED	National Academy Member
Ki Wook Kim	0.07	
FTE Equivalent:	**0.07**	
Total Number:	**1**	

Names of Under Graduate students supported

NAME	PERCENT_SUPPORTED
FTE Equivalent:	
Total Number:	

Student Metrics

This section only applies to graduating undergraduates supported by this agreement in this reporting period

The number of undergraduates funded by this agreement who graduated during this period: 0.00

The number of undergraduates funded by this agreement who graduated during this period with a degree in science, mathematics, engineering, or technology fields:...... 0.00

The number of undergraduates funded by your agreement who graduated during this period and will continue to pursue a graduate or Ph.D. degree in science, mathematics, engineering, or technology fields:...... 0.00

Number of graduating undergraduates who achieved a 3.5 GPA to 4.0 (4.0 max scale):...... 0.00

Number of graduating undergraduates funded by a DoD funded Center of Excellence grant for Education, Research and Engineering:...... 0.00

The number of undergraduates funded by your agreement who graduated during this period and intend to work for the Department of Defense 0.00

The number of undergraduates funded by your agreement who graduated during this period and will receive scholarships or fellowships for further studies in science, mathematics, engineering or technology fields:...... 0.00

Names of Personnel receiving masters degrees

NAME

Total Number:

Names of personnel receiving PHDs

NAME

Total Number:

Names of other research staff

NAME	PERCENT_SUPPORTED
FTE Equivalent:	
Total Number:	

Sub Contractors (DD882)

Inventions (DD882)

Scientific Progress

Technology Transfer

Scientific Progress and Accomplishments

This research project pursued theoretical understanding of magnetic correlation in both the graphene and the newly emerging topological insulator based structures for potential device applications. Highlights include new device concepts for low-power magnetic switch with all electrical control, electrically modulated domain wall transfer, sensitive room-temperature detection of magnetic fields and THz/far-IR radiation, as well as an unusual giant magnetoresistive effect with a single magnetic barrier.

- We explored electrically controlled magnetic correlation in graphene based composite structures for potential device applications. Prompted by recent studies of the Kondo effect in graphene that illustrated a strong influence of localized spin moments (associated with vacancies) through their interaction with the itinerant electrons, we investigated the role of carrier-impurity exchange interaction in formation of magnetic phase states of graphene with vacancies and modification in graphene-based composite structures. Our theoretical analysis based on a mean field model showed that the electronic free energy in graphene is indeed modified by the exchange interaction with localized spin moments. The calculation further revealed that at low temperatures a phase transition to antiferromagnetic (AF) ordering of the magnetization occurs, which in turn lead to a band gap opening between the conduction and valence bands. The AF phase transfer also has a significant impact on magnetic properties of graphene based composite structures. In a sandwich configuration (i.e., graphene placed in the middle), graphene is found to mediate the exchange bias field between two adjacent ferromagnetic (FM) layers. Accordingly, the exchange bias field can be altered and even reversed by modulating electronic properties of graphene. Our preliminary results thus far demonstrated that a sufficiently large vacancy concentration induces antiparallel alignment of two FM magnetizations, while the direction of the exchange bias field flips at low concentrations favoring parallel alignment. At the same time, it was shown that a high electron density can drastically enhance the strength of the effective magnetic field. As the electron density in graphene can be manipulated electrically, the observed phenomenon may be utilized for electrically controlled magnetization reversal.
- We proposed and theoretically analyzed a new scheme for electrically controlled magnetic domain wall transfer in graphene-ferromagnet composite structures for magnetoelectronic device applications. Unlike the conventional approach based on spin-momentum transfer, the proposed concept takes advantage of the proximity exchange interaction between graphene electrons and magnetic ions in a ferromagnetic layer and the fact that the resulting "effective magnetic field" can be manipulated by merely modulating the electron density in graphene (i.e., no current flow/low power). Figure 1 shows the structure under consideration. It consists of monolayer graphene and a thin ferromagnetic insulator (FMI) on top. Then, two ferromagnetic metallic

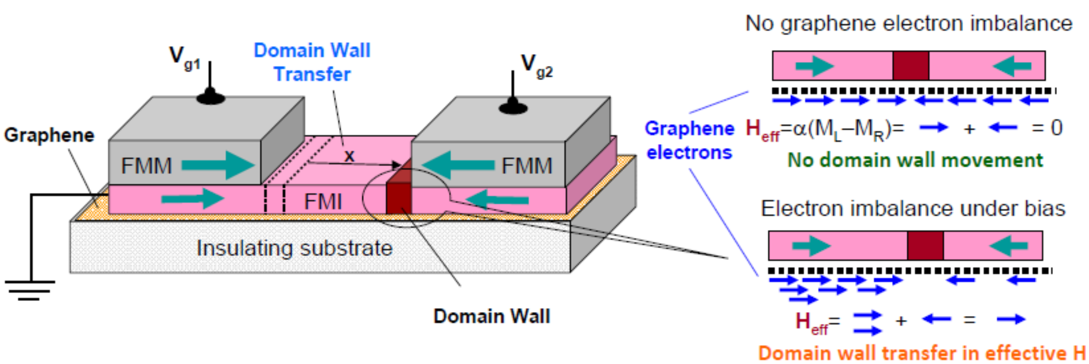

Figure 1: Schematic illustration of domain wall transfer in the proposed graphene-ferromagnet structure. The imbalance in the graphene electron density (thus, net electron spin polarization) can cause an effective magnetic field in the ferromagnetic insulator layer (FMI) in contact via the exchange interaction. Hence, the domain wall can be moved toward the desired direction by controlling the gate biases, V_{g1} and V_{g2}.

(FMM) gates are formed with opposite magnetization directions. Magnetization of the FMI regions (i.e., domains) under the gate is determined by that of the respective FMM. Hence, a domain wall is formed in the middle region. Electrical biases applied to the gate can vary the electronic density in the corresponding area of graphene. At zero (or identical) bias, both magnetic domains (with opposite magnetization) contribute equally to the spin polarization of graphene electrons in the middle region, negating each other. On the other hand, net polarization can be achieved when an imbalance in the electron density occurs (i.e., via uneven gate biases). This nonzero spin polarization acts like an effective magnetic field H_{eff} in the ungated region of FMI, pushing the domain wall in the desired direction. A preliminary estimate shows that H_{eff} of approx. 1000 oe is attainable for the bias of μ_L-μ_R=200 meV (between two gated regions in graphene) and the FMI thickness of 1 nm, sufficiently large for realistic application. More detailed calculations are currently under way for accurate evaluation of the performance under realistic device conditions.

- We proposed and theoretically analyzed a graphene-based spin capacitor and its potential application to sensitive magnetic field detection at room temperature. The main characteristic feature of the proposed device is essentially similar to a spin FET using graphene ribbon as the channel but has only one ferromagnetic (FM) terminal (say, source or drain). Thus, it forms a capacitor between the gate and FM S/D contacts, which stores not only the electric charge under an appropriate electrical bias but also spin polarization of those electrons (injected from the FM S/M) and their time evolution. Unlike the electric charge, spin polarization changes as a function of time due to spin precession in a magnetic field and decoherence. As soon as the sign of the gate bias is reversed, the electrons leave the capacitor with rotated and reduced spin polarization which, combined with the fixed magnetization of the FM S/D, can be detected electrically via the terminal current. Since the degree of rotation and decay is determined by the duration of electron spin exposure to the magnetic field as well as the strength and orientation of magnetic field in reference to the injected electron spin polarization, a series of measurements with differing detector orientation and exposure time can determine the strength and direction of the external (i.e., target) magnetic field. The proposed device provides a significant advantage over the schemes based on transient two-terminal measurement as the output signal is immune from the spin dephasing originating from the dispersion in transit time. Assuming the frequency measurement error of the order of a few per cent and the electron spin relaxation time of 100 ns at 300 K (a conservative choice), this device is capable of detecting magnetic fields in the 10 mOe (i.e., 0.01 Oe) range at the ambient temperature without involving any cooling or special equipment.
- We investigated transverse redistributions of the electrons and holes in intrinsic graphene ribbons under the influence of crossed electric and magnetic fields, i.e., the magnetoconcentration effect. Specifically, we examined a monolayer graphene ribbon in the xy plane confined by the width W in the y direction; the electric **E** and magnetic **H** fields were applied along the x and z axes, respectively. The magnetic field was considered to be classically strong, i.e., it does not lead to the formation of Landau levels. When the ribbon width exceeds the electron hole de Broglie wavelength, quantization of the electron and hole energy spectrum is not essential and can be disregarded. The ribbon length in the x direction was assumed to provide the longest dimension allowing a 1D treatment of the problem, with the inhomogeneity along the y axis. The nonequilibrium inhomogeneous redistribution of carriers under the influence of the fields was obtained by solving the Boltzmann kinetic equation within the local quasiequilibrium approximation. The calculation illustrates that an effective control of the carriers can be achieved from deep depletion to accumulation modes depending on the properties of the ribbon edges, provided electron-hole recombination/generation rates at the edges are different from those inside the ribbon. The current-voltage characteristics reflect the behavior of the carrier redistributions across the ribbon. Accordingly, interesting effects such as population inversion (accumulation

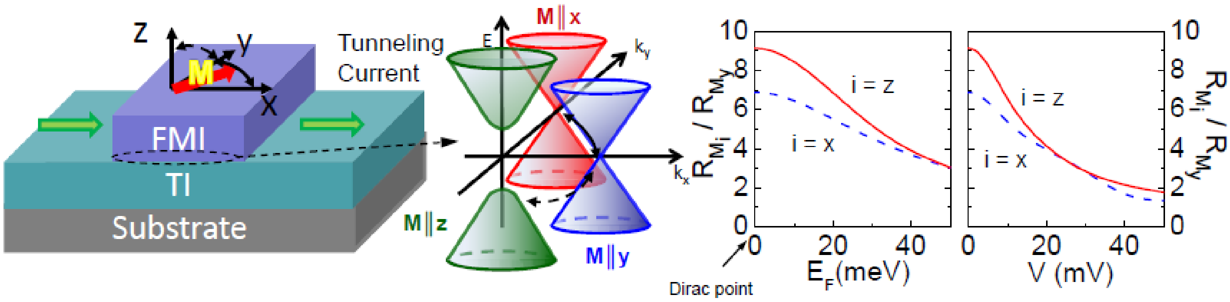

Figure 2: (Left) Schematic illustration of the proposed TI/FMI system. (Middle) Rotation of the magnetization direction around the x (**M**‖y ↔ **M**‖z) or the z (**M**‖y ↔ **M**‖x) axes leads to modification in the bandstructure of the TI surface states (i.e., the tunneling barrier). (Right) Relative ratio of surface resistance as a function of Fermi energy and gate bias when the magnetization rotates 90° (**M**‖y → **M**‖{x,z}). The solid lines represent the values of $R_{\mathbf{M}\|z}/R_{\mathbf{M}\|y}$, while the dashed lines show those of $R_{\mathbf{M}\|x}/R_{\mathbf{M}\|y}$. A 30-nm FMI strip is considered at room temperature along with the exchange energy of 40 meV at the TI/FMI interface. Apparently, the surface channel resistance can change by nearly an order of magnitude through magnetization rotation.

mode) and induced transparency (depletion mode) may be achieved with potential implication to THz device applications.

- We extended the investigation of the proximity exchange interaction with ferromagnetic materials to another 2D electronic system, namely the surface states of a 3D topological insulator (TI). One crucial property of TIs is the very strong spin-orbit coupling that leads to the intimately intertwined magnetic interactions and electron orbital motions. Consequently, the focus was to exploit the feasibility of current control via spin magnetic effects. The test structure has a single FMI layer of finite width placed on a TI, whose magnetization orientation can be varied in reference to the incident current (see Fig. 2). Accordingly, the FMI layer in this design was envisioned to play the role of a tunnel barrier with a controlling knob (analogous to the gate). A theoretical analysis based on a tight-binding effective Hamiltonian demonstrated that a single ferromagnetic barrier with variable magnetization can indeed modulate the electrical current on the surface of a TI. Numerical estimates suggest that the channel resistance change of a few hundred percent may be achieved even at room temperature in the ballistic tunnel transport regime. The key condition is sufficient exchange coupling at the TI/FMI interface (> 20 meV), which appears practically attainable in the current technology. The induced change in the channel resistance as a function of the magnetization direction is expected to find realistic applications in both memory and logic devices.
- We applied the magneto-electric phenomena of TI surface states to the prediction/discovery of a linear photo-galvanic effect (PGE) at the TI/FMI interface [Fig. 3(a)]. Theoretical analysis indicates that dc flow of photo-excited carriers can be achieved by asymmetric TI surface band distortion under the influence of both symmetry-breaking proximate exchange interaction and spin-independent parabolicity of the dispersion law [Fig. 3(b,c)]. Subsequent momentum-space imbalance in the generation of electron/hole pairs (with opposite group velocities) results in net photocurrent whose direction is also controlled by the chemical potential. The proposed PGE correlates with light absorption in a wide frequency range (from a few meV to hundreds of meV) and possesses a pronounced resonant response to the photon energy about two times the TI chemical potential shift from the Dirac point – thus, tunable by an electrical gate bias (Fig. 4). Moreover, it is invariant to the sign of light circular polarization (i.e., no polarized radiation necessary). The exceptionally strong peak photocurrent of the order of tens of μA/cm may be achieved with the illumination power of 1 W/cm2 in the THz range (estimate based on Bi_2Se_3; see Fig. 4). The proposed TI/FMI structures may have significant advantages over the conventional devices in the detection of long-wavelength photons beyond the thermal noise limit.

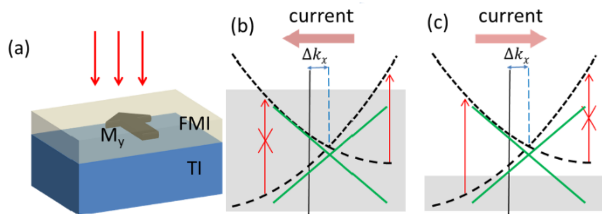

Figure 3: Schematic illustration of proposed PGE. (a) TI/FMI hybrid structure. The red arrows indicate the incident IR radiation; the block arrow denotes the magnetization direction. (b,c) Photocurrent generation in n-type and p-type TIs, respectively. The solid lines illustrate only the proximity effect on the TI surface band structure (i.e., the displacement of Δk_x). The dashed lines show the same bands when the quadratic term is also included. The shaded area denotes the filled electronic states.

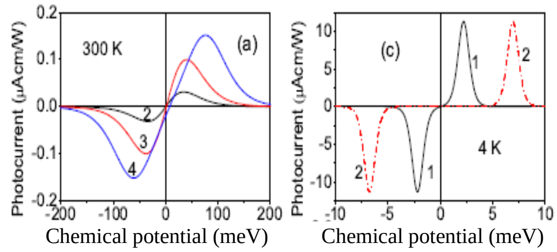

Figure 4: Photocurrent density calculated at (a) 300 K and (c) 4K versus the chemical potential with different excitation energies: 4.4 meV, 13.7 meV, 50 meV, and 135 meV for curves 1, 2, 3 and 4, respectively. All calculations assume the relaxation time of 1 ps and the proximate exchange energy of 40 meV along with material parameters of Bi_2Se_3.